

Spin Transitions in Graphene Butterflies at an Integer Filling Factor

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Recent experiments on the role of electron-electron interactions in fractal Dirac systems have revealed a host of interesting effects, in particular, the unique nature of the magnetic field dependence of butterfly gaps in graphene. The novel gap structure observed in the integer quantum Hall effect is quite intriguing [Nat. Phys. **10**, 525 (2014)], where one observes a suppression of the ferromagnetic state at one value of the commensurable flux but a reentrant ferromagnetic state at another. Our present work that includes the interplay between the electron-electron interaction and the periodic potential, explains the underlying physical processes that can lead to such a unique behavior of the butterfly gaps in that system where spin flip transitions are involved in the ground state.

The fascinating dynamics of Dirac fermions in graphene has been exhaustively studied in recent years [2–4]. Coulomb interaction between Dirac fermions [5], in particular, in the presence of a strong perpendicular magnetic field has resulted in the fractional quantum Hall states in monolayer [6] and bilayer graphene [7], which have also been experimentally observed [8]. Graphene placed on boron nitride with a twist displays fractal butterflies [9, 10] of Dirac fermions [11], when subjected to a perpendicular magnetic field. After the exciting discovery of the fractal butterflies in graphene [12–14] more recent theoretical [15] and experimental [16] studies have focused on the influence of electron-electron interactions on the butterfly gaps. Given the intricacies of these gaps, the interaction effects are more complex in the integer and fractional quantum Hall effect regime, where one observes an interplay between the quantum Hall effect gap and the Hofstadter gap [17]. In studying the interaction effects in the integer quantum Hall effect regime, Yu et al. [16] employed capacitance spectroscopy to explore the ‘Hofstadter minigaps’ for zero and integer filling factors. Their results for the energy gaps at filling factors $\nu = 0, \pm 1$ ($\nu = n\phi_0/B$, n is the particle density and ϕ_0 is the flux quantum) showed very unusual magnetic field dependence. In the low magnetic field region, the $\nu = 0$ gap rises *linearly* with B and saturates near the magnetic flux value $\phi = \phi_0/2$, but exhibits a minimum at $\phi = \phi_0$. In these two regimes, the gap deviates significantly from the Coulomb energy thereby indicating that the transitions across the gap from the ground state do not necessarily involve the particle charge alone.

By employing the magnetic translation group algebra [18] in the quantum Hall effect regime [17, 19–21], we have analyzed the magnetic field dependence of the $\nu = 0$ butterfly gaps. Our results reveal that the observed gap structure involve spin flip transitions in the ground state, as explained below. We consider graphene in an external periodic potential [15, 22–24]

$$V(x, y) = V_0[\cos(q_x x) + \cos(q_y y)], \quad (1)$$

where V_0 is the amplitude of the periodic potential and $q_x = q_y = q_0 = 2\pi/a_0$, where a_0 is the period of the

external potential. Then the many-body Hamiltonian is

$$\mathcal{H} = \sum_i^{N_e} [\mathcal{H}_B^i + V(x_i, y_i)] + \frac{1}{2} \sum_{i \neq j}^{N_e} V_{ij} \quad (2)$$

where \mathcal{H}_B^i is the Hamiltonian of an electron in graphene in a perpendicular magnetic field and the last term is the Coulomb interaction. The electron energy spectrum of graphene has twofold valley and twofold spin degeneracy in the absence of an external magnetic field, the periodic potential and the interaction between the electrons. It is well known [2, 3] that for magnetic fields that are presently accessible in the experiments, the conservation of the SU(2) valley symmetry in the presence of the Coulomb interaction is a fully justified approximation. We therefore employ this approximation in our studies. In order for the external periodic potential to break the SU(2) valley symmetry, the scattering process will require momentum transfer comparable to the value of the difference of momentum between the two valleys. The period of the external potential accessible in the experiment for the moiré superlattice is much bigger than the graphene lattice constant. Therefore the probability for such a momentum transfer processes is exponentially small and can be disregarded. In order to investigate what kind of state of total valley and real spin is favored by the system in our calculations, we consider both the spin and valley degrees of freedom of the electron system, where the spin degeneracy is lifted due to the Zeeman effect, while in the approximation described above there is no term in the Hamiltonian (2) to lift the valley degeneracy. The single-particle Hamiltonian \mathcal{H}_B is then written as [2–4]

$$\mathcal{H}_B = \xi v_F \begin{pmatrix} 0 & \pi_- \\ \pi_+ & 0 \end{pmatrix} + \frac{1}{2} g_e \mu_B B \sigma_z, \quad (3)$$

where $\pi_{\pm} = \pi_x \pm i\pi_y$, $\boldsymbol{\pi} = \mathbf{p} + e\mathbf{A}/c$, \mathbf{p} is the two-dimensional electron momentum, $\mathbf{A} = (0, Bx, 0)$ is the vector potential, $v_F \approx 10^6$ m/s is the Fermi velocity in graphene and the last term is the electron Zeeman energy. The ξ is the valley index: $\xi = 1$ for valley K and $\xi = -1$ for valley K' . The honeycomb lattice of graphene consists of two sublattices A and B and the two component

wave functions corresponding to the Hamiltonian (3) can be expressed as $(\psi_A, \psi_B)^T$ for valley K and $(\psi_B, \psi_A)^T$ for valley K' , where ψ_A and ψ_B are wave functions of sublattices A and B, respectively. The eigenfunction of the Hamiltonian (3) for both K and K' valley can be written in the form [2–4]

$$\Psi_{n,j} = C_n \begin{pmatrix} \text{sgn}(n)(-i)\varphi_{|n|-1,j} \\ \varphi_{|n|,j} \end{pmatrix}, \quad (4)$$

where $C_n = 1$ for $n = 0$ and $C_n = 1/\sqrt{2}$ for $n \neq 0$, $\text{sgn}(n) = 1$ for $n > 0$, $\text{sgn}(n) = 0$ for $n = 0$, and $\text{sgn}(n) = -1$ for $n < 0$. Here $\varphi_{n,j}$ is the electron wave function in the n -th LL with the parabolic dispersion, taking into account the periodic boundary conditions (PBC) [21, 25]. The eigenvalues of Hamiltonian (3) corresponding to the eigenvectors (4) for both valleys K and K' are $\epsilon_n = \text{sgn}(n)\hbar\omega_B\sqrt{|n|}$, where $\omega_B = \sqrt{2}v_F/\ell_0$, $\ell_0 = \sqrt{\hbar c/eB}$ is the magnetic length.

We consider a system of finite number N_e of electrons in a toroidal geometry, i.e., the size of the system is $L_x = M_x a_0$ and $L_y = M_y a_0$ (M_x and M_y are integers) and apply PBC in order to eliminate the boundary effects. Defining the parameter $\alpha = \phi_0/\phi$, where $\phi = Ba_0^2$ is the magnetic flux through the unit cell of the periodic potential and $\phi_0 = hc/e$ the flux quantum, we have

$$\frac{N_s}{M_x M_y} = \frac{1}{\alpha}, \quad (5)$$

where N_s describes the LL degeneracy for each value of the spin and valley index. In this work we consider the filling factor of $\nu = 0$, which means that the number of electrons in the zeroth LL is $N_e = 2N_s$, because of the fourfold degeneracy of each LL in graphene. The procedure of constructing the Hamiltonian matrix in the basis of the many-body states $|j_1, j_2, \dots, j_{N_e}\rangle$ (besides j_i , each single-particle state is characterized by the LL, spin and valley indices which are not shown, but are implicitly assumed to be included in the indices j_i) constructed from the single-particle eigenvectors (4). Diagonalization of this matrix follows the procedure outlined in [17]. It was previously shown [17] that the center of mass (CM) translations $T^{\text{CM}}(\mathbf{a}_p)$ with the translation vector $\mathbf{a}_p = m\beta_1 a_0 \hat{\mathbf{x}} + n\beta_2 a_0 \hat{\mathbf{y}}$ can be used to characterize the eigenstates of Hamiltonian (2), where the eigenvalues of the CM translation operator have the form $e^{2\pi i(\beta_1 ms/M_x + \beta_2 nt/M_y)}$. Here, m and n are integers which define the translation vector, β_1 and β_2 are integers which characterize the degeneracy of the many-body system and can be determined from the relation $N_e\beta_1\beta_2/\alpha = \pm 1, \pm 2, \dots$, s and t are also integers which are defined modulo M_x/β_1 and M_y/β_2 respectively and describe the eigenvalues of the CM translation operator. The CM translations allow us to divide the basis states into equivalence classes and transfer the complete Hamiltonian into block diagonal form where each block is diagonalized separately and also to characterize each

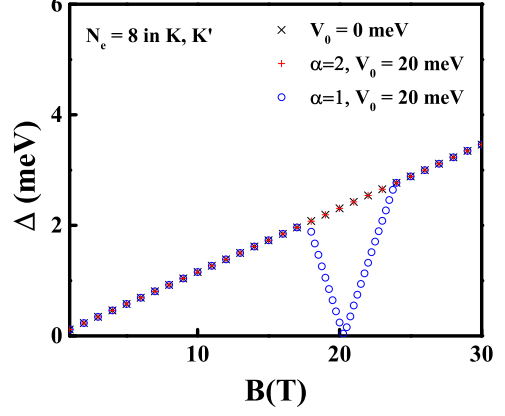


FIG. 1: The gap between ground and first excited state of an eight-electron system in two valleys versus the magnetic field B for two values of α , with $V_0 = 20$ meV and without the external periodic potential.

eigenvalue of the Hamiltonian with the appropriate CM momentum. Therefore using the CM translation analysis the Hamiltonian matrix can be approximately divided into $M_x M_y / \beta_1 \beta_2$ separate blocks.

In what follows we consider the two cases $\alpha = 1$ and $\alpha = 2$. We then choose the system size based on the condition (5) and the number of electrons. After comparing the results for small system sizes for the cases with and without the inclusion of the contribution of higher LLs, in what follows we disregard the LL mixing and present all the results for the $n = 0$ LL. Here we present the results for three system sizes, $N_e = 8$ taking into account both K and K' valleys, $N_e = 6$ and $N_e = 8$ taking into account only the K valley. For $N_e = 8$ in two valleys the system size is $M_x = 2$ and $M_y = 2$ for $\alpha = 1$, and $M_x = 4$ and $M_y = 2$ for $\alpha = 2$. For $N_e = 6$ in one valley the system size is $M_x = 3$ and $M_y = 2$ for $\alpha = 1$, and $M_x = 6$ and $M_y = 2$ for $\alpha = 2$, and for $N_e = 8$ in one valley the system size is $M_x = 4$ and $M_y = 2$ for $\alpha = 1$, and $M_x = 4$ and $M_y = 4$ for $\alpha = 2$. In order to investigate the magnetic field dependence of the gap we fix the value of α and change the magnetic field B and the period of the periodic potential simultaneously.

In Fig. 1 the dependence of the gap between the ground state and the first excited state on the magnetic field strength is presented for $N_e = 8$ electrons in K and K' valleys for $\alpha = 1$ and for $\alpha = 2$ with $V_0 = 20$ meV and without the periodic potential. In the absence of the periodic potential the change of α results only in the change of geometry of the system and therefore does not have any contribution in the gap value and in the figures the $V_0 = 0$ case is presented without the indication of the value of α . For $V_0 = 0$ and for all values of the magnetic field the ground state corresponds to four electrons in each valley, all electrons having their spins in the opposite direction to the magnetic field. As for the excited

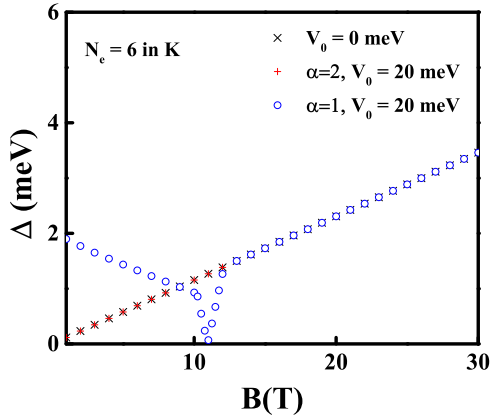


FIG. 2: Same as in Fig. 1, but for the system of six electrons. Only the K valley is considered in this case.

state, it corresponds to the spin flip of one electron from eight electrons and the cases when the spin flip electron is located in the same valley as the partially filled spin down state with three electrons or they are located in two different valleys are degenerate. It should be also noted that there is no momentum transfer in this transition from the ground state to the excited state described above and the gap is equal to the Zeeman energy of the spin flip. Therefore for $V_0 = 0$ the electron-electron interaction does not have any contribution in the lowest gap of the system. As can be seen from Fig. 1, surprisingly the situation remains the same for $\alpha = 2$ and $V_0 = 20$ meV.

As for the case of $\alpha = 1$ and $V_0 = 20$ meV the situation is different. In the magnetic field region up to 18 Tesla the total spin of the ground state is $S_z = -2$ and there are four electrons in each valley. This corresponds to the case when in each valley three electrons have spin down and one electron spin up. Therefore, the periodic potential changes the ground state of the system from the fully spin polarized state to the spin partially polarized state in this case. As for the excited state the total spin is $S_z = -1$, which again means one additional spin flip and again the level is degenerate with respect to the exchange of spin up parts between the valleys. In Fig. 1 the gap is again equal to the Zeeman energy of the spin flip. At $B \approx 18$ Tesla there is a crossing between the first and the second excited states and up to $B = 20$ Tesla the first excited state total spin is $S_z = -4$, and the gap correspond to the transition between the states with double spin flip from the spin up to down and is again equal to the Zeeman energy of that transition. At $B \approx 20$ Tesla there is a crossing between the ground and the first excited state, and after that the ground state is the state with total spin $S_z = -4$. The first excited state is the state with total spin $S_z = -2$ up to 24 Tesla and the state with total spin $S_z = -3$ (which is again degenerate due to the different configurations of the spin up states between two valleys)

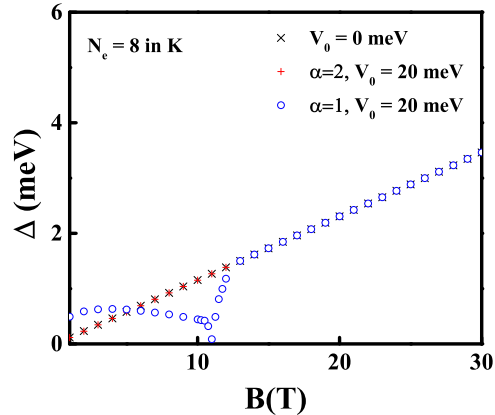


FIG. 3: Same as in Fig. 2 but for the system of eight electrons. Only the K valley is considered in this case.

afterwards. For these two parts, the gap is again equal only to the Zeeman energy for the appropriate transition. It should be noted that for the range of magnetic fields considered in this case both the ground and the excited state are described by the total momentum equal to zero and there is no momentum transfer in these transitions. Although the gap energy for this case is always equal to the Zeeman energy of the appropriate transition, the gap structure shown for $\alpha = 1$ and $V_0 = 20$ meV is not the single-particle effect. Both the electron-electron interaction and the periodic potential are essential for the system to deviate from the ferromagnetic state at low magnetic fields and afterwards for the observation of the transition from a spin partially polarized state to the fully polarized spin state by increasing the magnetic field.

In Fig. 2 and Fig. 3 the dependence of the gap between the ground state and the first excited state on the magnetic field strength are presented for $N_e = 6$ and $N_e = 8$ electrons for $\alpha = 1$ and $\alpha = 2$ with $V_0 = 20$ meV and without the periodic potential. Only the K valley is considered in these cases. The cases of $V_0 = 0$ and $V_0 = 20$ meV with $\alpha = 2$ show the same behavior as for the case of $N_e = 8$ electrons in K and K' valleys shown in Fig. 1. The ground state is the ferromagnetic state for all values of the magnetic field and the gap corresponds to the transition to the excited state with a single spin flip. For the case of $\alpha = 1$ and $V_0 = 20$ meV the situation is different. At small magnetic fields (up to 10 Tesla) the ground state total spin is equal to $S_z = 0$ (spin unpolarized state) and the transition corresponds to the excited state with total spin equal to $S_z = -1$. It should be noted that in addition to the spin flip, there is also the momentum transfer in these transitions, because the first excited state is characterized by a nonzero momentum up to 10 Tesla. Therefore these transitions correspond to collective excitations and the gap energy is comprised both the Zeeman term plus the $\Delta \propto \sqrt{B}$ term. This

structure is clearly visible for the $N_e = 8$ electron case, but for the $N_e = 6$ electron case the collective nature of the excitation is completely suppressed by the Zeeman term for magnetic fields considered in Fig. 2. In Fig. 2 and Fig. 3, starting with $B = 10$ Tesla the structure of the transition is the same as for the case of $N_e = 8$ electrons in K and K' valleys shown in Fig. 1. There are several crossings between the low-lying excited states and the ground states of the system passes from the spin unpolarized ground state to the ferromagnetic (fully polarized) state. In the ferromagnetic regime the gap again corresponds to the spin flip and is equal to the Zeeman energy of that flip.

We now use the features observed in this work to interpret the result shown in Fig. 4 of Ref. [16] for the filling factor $\nu = 0$. For magnetic fields up to 20 Tesla, due to the valley anisotropic terms [26] and also due to the spin unpolarized state observed for low magnetic fields in our work, presumably the system is in a spin unpolarized state. The almost linear dependence of the gap at magnetic fields up to around 5 Tesla and the \sqrt{B} dependence for magnetic fields between 5-20 Tesla indicates that the excitations have both spin flip and the momentum transfer component (collective excitation). Based on these observations it can be assumed that there is a competition between these two components and that for up to 5 Tesla magnetic fields the Zeeman contribution is dominant in the gap energy, whereas in the range of magnetic fields 5-20 Tesla the electron-electron interaction contribution is dominant. The lowering of the gap

in the region close to $\alpha = 1$, then almost linear dependence after around 28 Tesla and the absence of similar features for $\alpha = 2$ closely resembles the results obtained in our paper. We therefore suggest that this behavior indicates that there is a phase transition around $\alpha = 1$, i.e., there is a transition from the spin unpolarized state to the partially or fully spin polarized state.

In conclusion, we have considered the influence of the periodic potential due to the moiré lattice on the dependence of the energy gap on the magnetic field for filling factor $\nu = 0$ in graphene using the exact diagonalization scheme. We have considered three cases: $N_e = 8$ electrons in K and K' valley, $N_e = 6$ and $N_e = 8$ electrons located only in the K valley and disregarding the contribution of the K' valley. In all cases we find that for $\alpha = 1$, inclusion of the periodic potential and the electron-electron interactions results in unpolarized or partially polarized ground state at lower magnetic fields and a transition to the fully polarized ground state by increasing the magnetic field. This behavior is not observed for the $\alpha = 2$ case, where we only observe the ferromagnetic state for all values of the magnetic field. The similarity between the results obtained here and reported in Fig. 4 of Ref. [16] was analyzed and a possible explanation was presented on the behavior of the dependence of the gap on the perpendicular magnetic field observed in that work.

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